

**PATENT APPLICATION**

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of

Docket No: Q95621

Masanobu AIZAWA

Appln. No.: 10/590,234

Group Art Unit: 1797

Confirmation No.: 6972

Examiner: Benjamin M. Kurtz

Filed: August 22, 2006

For: SEPARATION MEMBRANE

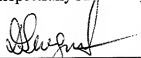
**SUBMISSION OF EXECUTED DECLARATION UNDER 37 C.F.R. §1.132**

Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

Submitted herewith is a copy of an executed Declaration Under 37 C.F.R. §1.132 signed  
by Mr. Junji Saito.

Respectfully submitted,



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CUSTOMER NUMBER

Date: February 23, 2011

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**DECLARATION UNDER 37 C.F.R. § 1.132**

Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Junji Saito, hereby declare and state:

THAT I am a citizen of Japan;

THAT I have received a Bachelor's Degree in Applied Chemistry in 1971 from the  
Faculty of Engineering, Hokkaido University;

THAT I have received a Master's Degree in Applied Chemistry in 1973 from the Faculty  
of Engineering, Hokkaido University;

THAT I was employed by Mitsubishi Chemical Corporation from April, 1973 to March,  
2003;

THAT I was employed by Bussan Nanotech Research Institute, Inc. from April, 2003 to  
June, 2008;

THAT I have been employed by Mitsubishi Chemical Corporation since July, 2008;

THAT I have been engaged in the research and development of zeolites from April, 2003 onwards;

THAT I am familiar with the Office Action dated September 28, 2010;

THAT I am aware that Claims 1, 3-5 and 9-15 were rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Lai et al. (U.S. Patent No. 5,871,650) and Verduijn et al. (U.S. Patent No. 6,090,289) in view of Goldsmith et al. (U.S. Patent No. 5,221,484); and, Claims 7 and 8 were rejected under 35 U.S.C. § 103(a) as allegedly being unpatentable over Lai, Verduijn, Goldsmith, and further in view of Mover et al. (U.S. Patent No. 5,198,007); and,

I submit the present Declaration in support of the patentability of the invention described in the above-identified application over Lai. My Declaration provides comparative evidence which shows that the separation membrane described in Lai can not achieve both high separation capability and high permeation rate, and thus, the separation membrane of the presently claimed invention shows unexpectedly superior results over the separation membrane described in Lai in terms of nitrogen gas permeation rate, flux  $Q$  and separation factor  $\alpha$ .

#### **EXPERIMENTATION**

The experiment described in this Declaration is similar to the experiment described in Example 1 at paragraphs [0048]-[0054] of the specification.

First, sintering auxiliary agent powder, which consists of  $MgO$  and  $CaCO_3$ , and water were mixed together using a ball mill. Then, high purity alumina powder and methyl cellulose type binder were provided in order to knead them with the mixture obtained in the previous step, and the resultant kneaded mixture was subjected to extrusion molding in order to mold a primer

tube, which was followed by drying the primer tube and degreasing the dried primer tube. The primer tube was then sintered. The primer tube made of ceramic sintered body of which the main ingredient is alumina was prepared. The mean pore diameter of the base layer was 7  $\mu\text{m}$ , and the porosity was 40%. A foundation layer was formed on the outer surface of the primer tube. High purity alumina powder,  $\alpha$ -terpineol, ethanol, and ethyl cellulose type binder were blended in a weight ratio of 30:75:25:4 and stirred in order to prepare slurry. Into the obtained aluminum colloidal particle slurry, the aforementioned primer tube was dipped in order to attach the slurry on the outer surface of the primer tube. Then, the primer tube was dried, and sintered at a temperature of 500°C in order to form a foundation layer on the outer surface of the primer tube.

The porous substrates of Experiments 1-3, which comprised the primer tube, which was provided with a foundation layer on the outer surface thereof, was prepared. The mean pore diameter of the foundation layer of Experiment 1 was 0.17  $\mu\text{m}$ ; the mean pore diameter of the foundation layer of Experiment 2 was 0.07  $\mu\text{m}$ ; and the mean pore diameter of the foundation layer of Experiment 3 was 0.8  $\mu\text{m}$ . The porosity of the foundation layer, the thickness of the foundation layer and the nitrogen gas permeation rate of the porous substrate were measured. The results are shown in the Table below.

Minute particles of zeolite (diameter: 300 nm) were added and mixed to water, then they were stirred together in order to prepare a slurry having a concentration of 0.5% by weight. To this slurry the aforementioned porous substrate made of  $\alpha$ -alumina (outer diameter: 10 mm, inner diameter: 5mm, length: 13 cm) was dipped for 3 minutes, and then it was pulled up from the

slurry at a rate of about 0.2 cm/sec. The dipped porous substrate was then dried for 2 hours in a temperature controlled bath of 23°C, and for 16 hours in another temperature controlled bath of 40°C.

Hydrothermal reaction solution of pH 13 was prepared by mixing sodium silicate, aluminum hydroxide and distilled water so that the mole ratios of respective ingredients satisfied the conditions of  $\text{SiO}_2/\text{Al}_2\text{O}_3 = 2$ ,  $\text{Na}_2\text{O}/\text{SiO}_2 = 1$ , and  $\text{H}_2\text{O}/\text{Na}_2\text{O} = 75$ . The seed crystals attached porous substrate was dipped into this reaction solution and maintained therein for five hours at 100°C, resulting in the formation of a zeolite membrane on the surface of the porous substrate, i.e., the surface of the foundation layer.

A pervaporation (PV) testing apparatus as shown in Fig. 5 of the specification was assembled so as to evaluate the obtained separation membrane, i.e., the separation membrane in which the zeolite membrane was formed on the surface of the porous substrate, for separation performance. The PV testing apparatus included: a container 7 provided with a pipe 11 through which a feed liquid A is fed and a stirrer 12; a separation apparatus 8 installed in the inside of the container 7; a pipe 6 connected to the open end of the separation apparatus 8; and a vacuum pump 10 connected to the end of the pipe 6 via a liquid nitrogen trap 9. The separation apparatus 8 was made up of the above mentioned separation membrane, in which the zeolite membrane was formed on the surface of the porous substrate. The pipe 6 was equipped with a vacuum gauge 5 at some midpoint thereof. A feed liquid (the mass ratio of ethanol/water = 90/10) at 75°C was fed to the container 7 of the PV testing apparatus through the pipe 11 and suction was applied to the inside of the separation apparatus 8 with the vacuum pump 10 (the vacuum, as

measure by the vacuum gauge 5, was 10-1000 Pa). The liquid B having permeated the separation membrane 52 was trapped with the liquid nitrogen trap 9. The composition of the feed liquid A and the liquid B having permeated were measured by a gas chromatograph (GC-14B manufactured by Shimadzu Corporation), and the separation factor  $\alpha$  and the flux Q, which is the permeation rate of water, were determined. The results are shown in the Table below.

The separation membrane of Experiment 1 and the separation membrane of Experiment 2 are separation membranes mentioned in Lai because the mean pore diameter of the foundation layer of Experiment 1 was 0.17  $\mu\text{m}$ , the mean pore diameter of the foundation layer of Experiment 2 was 0.07  $\mu\text{m}$ , and Lai teaches that the pore diameter range of the mesoporous growth enhancing layer (GEL layer) is 20-2000 angstrom (0.002-0.2  $\mu\text{m}$ ). See, e.g., col. 4, lines 36-55 of Lai. The mean pore diameters of the foundation layers of Experiments 1 and 2 are not included in the presently claimed range of 0.4-1.2  $\mu\text{m}$ .

The separation membrane of Experiment 3 is the separation membrane of present Claim 1 because the mean pore diameter of the foundation layer of Experiment 3 was 0.8  $\mu\text{m}$ , which falls within the presently claimed range of 0.4-1.2  $\mu\text{m}$ .

	Experiment 1 (Lai)	Experiment 2 (Lai)	Experiment 3 (claim 1)
mean pore diameter of foundation layer [ $\mu\text{m}$ ]	0.17	0.07	0.8
porosity of foundation layer [%]	39	40	40
thickness of foundation layer [ $\mu\text{m}$ ]	50	60	50
nitrogen gas permeation rate of porous substrate [ $\text{m}^3/(\text{m}^2 \cdot \text{hr} \cdot \text{atm})$ ]	66	34	900
flux Q of separation membrane [ $\text{kg}/\text{m}^2 \cdot \text{hr}$ ]	3.7	2.2	7.9
separation factor $\alpha$ of separation membrane	2750	814	10900

Moreover, as show in the Table above, the nitrogen gas permeation rate, i.e.,  $66 \text{ m}^3/(\text{m}^2 \cdot \text{hr} \cdot \text{atm})$ , of the porous substrate of Experiment 1, and the nitrogen gas permeation rate, i.e.,  $34 \text{ m}^3/(\text{m}^2 \cdot \text{hr} \cdot \text{atm})$ , of the porous substrate of Experiment 2 is not included in the presently claimed range of  $200\text{-}7000 \text{ m}^3/(\text{m}^2 \cdot \text{hr} \cdot \text{atm})$ .

Finally, as shown in the Table above, the flux Q, i.e.,  $3.7 \text{ kg}/\text{m}^2 \cdot \text{hr}$ , of the separation membrane of Experiment 1, and the flux Q, i.e.,  $2.2 \text{ kg}/\text{m}^2 \cdot \text{hr}$ , of the separation membrane of Experiment 2, is not included in the presently claimed range of  $5.0 \text{ kg}/(\text{m}^2 \cdot \text{hr})$  or more.

Therefore, the separation membrane discussed in Lai cannot achieve both high separation capability and high permeation rate, as in the presently claimed invention.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these

DECLARATION UNDER 37 C.F.R. § 1.132  
U.S. Application No.: 10/590,234

Attorney Docket No.: Q95621

statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: February 4, 2011

Junji Saito  
Junji SAITO